In all of the above examples n is of sufficient value to produce a viscosity of 500 to 500,000 cs. at 25° C. While at least 80 mol percent of the R' groups must be methyl, it is preferable that at least 92.5 mol percent, of the R' groups be methyl groups. Also, while the viscosity can 5 range up to 500,000 cs., it is generally preferable that a lower viscosity material, for example a material not exceeding about 150,000 cs., be employed since such a material is more readily pourable.

For the purpose of this invention it is essential that 10 component (2) be employed in amount from 5 to 50% by weight of the combined weights of (1) and (2). When the compositions contain no filler, however, it is preferred that component (2) be employed in amount from 20 to 50% by weight. Composition (2) is a 15 copolymer of the three specified siloxane units. It is essential that the weight percent of the vinyl groups in said copolymers be from 1.5 to 3.5% based on the weight of (2). These percents are calculated based on the weight of the vinyl radical, that is 27. In order to obtain trans- 20 parent materials it is essential that the total trimethylsiloxane units and dimethylvinylsiloxane units relative to the SiO<sub>2</sub> units in (2) shall range from .6:1 to 1:1.

Copolymers (1) can be prepared by any of the conventional methods for preparing triorganosilyl-endblocked diorganopolysiloxanes. This can be done, for example, by cohydrolyzing and condensing the appropriate chloroand/or alkoxylsilanes, or by equilibrating the appropriate hexaorganodisiloxane with the appropriate diorganopolysiloxane in the presence of an alkaline or acid catalyst.

Copolymers (2) can be prepared by the cohydrolysis of the silanes of the formulae SiX<sub>4</sub>, Me<sub>3</sub>SiX and Me<sub>2</sub>ViSiX in the proper proportions to give the desired ratios. X is halogen or alkoxy radicals. Alternatively the copolymer can be prepared by the procedure described in U.S. Patent 2,676,182. This involves reacting a silica hydrosol with hexamethyldisiloxane or trimethylchlorosilane and with dimethylvinylchlorosilane or divinyltetramethyldisiloxane. The latter can be reacted with the silica hydrosol either simultaneously with or subsequently to reaction with the 40 trimethylchlorosilane or hexamethyldisiloxane. Preferably the vinyl constituent is reacted with the hydrosol subsequently to the trimethylsilyl constituent.

Another critical component of the composition of this invention is copolymer (3) which must be employed in amount sufficient that the ratio of mols of silicon-bonded hydrogen in the overall composition relative to the mols of silicon-bonded vinyl groups in (1) and (2) shall be within the range .75:1 to 1.5:1. Composition (3) must be compatible with (1) and (2) if a transparent material is to be obtained. Furthermore, in order to obtain an operative material the percent by weight of silicon-bonded hydrogen in (3) must be within the range of 0.1 to 1.7 inclusive percent by weight based on the weight of (3). In order for (3) to serve as a cross-linker for the sys- 55 tems, there must be at least three silicon-bonded hydrogens

Furthermore, those silicon atoms in (3) which have both hydrogen and hydrocarbon radicals bonded thereto, essentially all of these hydrocarbon radicals should be methyl. The term "essentially all" means that the presence of some PhHSiO groups is not precluded. However, if such groups are present there must also be at least three MeHSiO, Me2HSiO1/2 or HSiO3/2 groups per molecule.

Specific examples of (3) of which are within the scope of this invention are

 ${\rm HSi}({\rm OSiMe_2H})_2({\rm OSiMe_3}),\ {\rm Me_3Si}({\rm OSi}_1)_{10}{\rm OSiMe_3}\ {\rm and}\ {\rm Si}({\rm OSiMe_2H})_4$ 

copolymers of SiO<sub>2</sub>, Me<sub>2</sub>SiO and Me<sub>2</sub>HSiO<sub>1/2</sub>; copolymers of Me<sub>3</sub>SiO<sub>1/2</sub>, Me<sub>2</sub>HSiO<sub>1/2</sub>, MeHSiO and Me<sub>2</sub>SiO; cyclic (MeHSiO)x; copolymers of methyl hydrogen siloxane and phenylmethylsiloxane; copolymers of methyl hydro- 75 C. The material was cast around a steel hexagonal bar

gen siloxane, dimethylsiloxane and diphenylsiloxane and copolymers of methyl hydrogen siloxane and dimethylsiloxane.

Any finely divided form of platinum can be employed as the catalyst in this invention. This includes finely dispersed metallic platinum such as platinum dispersed on charcoal or other carriers, and soluble compounds of platinum such as chloroplatinic acid, or complexes of platinic chloride with olefins such as ethylene, propylene, butadiene, cyclohexene and the like. Preferably the platinum should be in soluble form.

The amount of platinum is not critical since it merely affects the rate of cure. However, for practical operation, it is preferred that the catalyst be employed in amount ranging from .5 to 20 parts per million based on the weight of (1), (2) and (3).

After the four ingredients have been mixed in any desirable manner, the composition is then cured. The composition will cure very slowly at room temperature, but for most applications it is preferred that the mixture be heated at a temperature of from 50 to 150° C. or above. This expedites the cure so that it will proceed in a matter of from less than one hour to several hours. Thus, it can be seen that one can regulate the time of cure widely by regulating the temperature and the concentration of the platinum catalyst.

If desired, fillers may be employed in the compositions of this invention. These fillers are used when optical clarity is not desired. The fillers operative herein include any of the fillers normally employed in organopolysiloxanes such as fume silica, aluminum silicate, quartz, calcium carbonate, potassium titanate, zirconium silicate, carbon blacks, and metal oxides such as alumina, zinc oxide, titania and ferric oxide. If desired, the fillers can be treated with organosilicon compounds such as chlorosilanes or alkoxysilanes so as to produce a hydrophobic surface. The latter treatment is particularly desirable with finely divided silicas, such as fume silicas or silica aerogels.

Optionally, one can also include polydimethylsiloxane fluids in the compositions of this invention as plasticizers.

The compositions of this invention are useful for electrical insulation and for the formation of cast articles. The compositions of this invention are also useful for making light-pipes.

The following examples are illustrative only and should not be construed as limiting the invention which is properly delineated in the appended claims. The following abbreviations are employed in this specification, Me is methyl, Ph is phenyl and Vi is vinyl. In the following examples all parts and percents are by weight unless otherwise stated. In all copolymers (2) in the following examples the ratios of total Me<sub>3</sub>SiO<sub>.5</sub> and Me<sub>2</sub>ViSiO<sub>.5</sub> to  $SiO_2$  units are in the range .6:1 to 1:1.

## Example 1

65% of (1) a 2,000 cs. viscosity siloxane of the for-

$$\begin{array}{c} {\rm PhMeViSiO} {{\rm (Me_2)}\atop{\rm SiO}}_{\rm nSiPhMeVi} \end{array}$$

was mixed with (2) 35% of a copolymer of SiO<sub>2</sub>, Me<sub>3</sub>SiO<sub>.5</sub> and Me<sub>2</sub>ViSiO<sub>.5</sub> containing 2.5% vinyl groups. 94.9 parts of the above mixture was mixed with (3) 65 5.1 parts of PhSi(OSiMe<sub>2</sub>H)<sub>3</sub> and 5 parts per million platinum added as chloroplatinic acid dissolved in 2ethylhexanol. The resulting mixture was cast into a slab 8 x 8 x 1/16 inch and cured one hour at 150° C. The resulting slab was clear and had the following properties:

Tensile strength \_\_\_\_\_p.s.i\_ 750 Percent elongation at break \_\_\_\_\_ 80

The above mixture had a viscosity of 4500 cs. at 25°